

***LONG-TERM NET IMPACTS OF AEROSOLS ON CLOUD AND  
PRECIPITATION***

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Aerosols have complex effects on cloud and precipitation that often offset each other, rendering an unknown net effect, especially on a long time scale. Using 10 years of ARM/SGP measurements, aerosol's net impacts emerge: rain frequency increasing with the ground-level aerosol concentration (CN) for clouds with large liquid water path (LWP), but decreasing for low LWP. As the CN increases, cloud thickness increase substantially (up to a factor of 2!) for low-base clouds (< 1 km) but little effect for high-base clouds (> 2 km). Using a simple conceptual model and a complex cloud resolving model, the findings are explained by the competition between the suppressed coalescence effect and the invigoration effect. Their relative strengths are dictated by cloud thickness and water content. This helps reconcile and generalize some controversial findings. The unprecedented signals of aerosols' net effects on cloud and precipitation have significant implications for hydrological cycling, anthropogenic forcing and policy-making.

## Background

Isolating and quantifying the impact of natural and anthropogenic aerosols on the Earth's hydrological cycle has been a major challenge in climate sciences (1). This challenge exists because aerosols have very complex radiative, thermodynamic and microphysical effects on clouds and precipitation (2) and their effects depend upon ever-changing meteorological conditions. The effects of aerosols on clouds and precipitation may work in harmony with or against each other, leading to seemingly controversial findings (3). For instance, aerosols were shown 50-y ago to suppress rainfall by reducing cloud droplet size and by slowing the conversion of cloud droplets into raindrops (4). Much later it was suggested that aerosols can also enhance rainfall from deep convective clouds by initially inhibiting precipitation at lower altitudes, so that the cloud water ascends to above the freezing level and creates ice hydrometeors. The additional released latent heat of freezing invigorates and deepens the convective clouds and intensifies the precipitation (5, 6). This hypothesis is consistent with cloud-resolving model simulations (7, 8). However, theoretical considerations and model simulations showed that in addition to aerosols, relative humidity, wind shear, radiative effects and other factors all play important roles in determining if a cloud is invigorated, unchanged or suppressed (3, 6, 9-11). The lack of knowledge of their relative importance, their interactions and the frequency of occurrence result in a *totally unpredictable net effect on a long-term basis*, which is of most concern to the earth's climate.

## Data

Since its inception in late 1980s (12, 13), the Department of Energy's Atmospheric Radiation Measurements (ARM, now the Atmospheric Science Research) has provided the most extensive, accurate and long-term observations attempting to understand and parameterize atmospheric processes in climate models. Among others, the ARM has developed and operated a suite of state-of-the-art passive and active instruments measuring cloud hydrometers and geometry (14, 15). Employed in this study are the continuous observations of cloud, precipitation and atmospheric variables acquired at the

Central Facility (CF) site over the SGP. They include aerosol number concentration (CN), cloud liquid water path (LWP), cloud top and base heights, precipitation and meteorological variables. The most unique data that are rarely available elsewhere are the cloud boundaries obtained from the ARSCL (Active Remote Sensing of Clouds). ARSCL is a value-added product derived for the period from 1999 to 2008 by combining data from millimeter cloud radars, laser ceilometers, microwave radiometers, and micropulse lidars (15). Two sets of precipitation data of complimentary merits are utilized that were acquired by the Surface Meteorological Observation System Instruments (SMOS) and the Carbon dioxide Flux Measurement System (CO2FLX). Both used tipping bucket rain gauge. SMOS data are available over the 10-y period, while the CO2FLX data are from 2003 to present. The minimum value in the 30-min data as used in this study is 0.254 mm from the SMOS, and 0.1 mm from the CO2FLX. Because of the higher initial detection value, the SMOS only detected 58% of rain events as the CO2FLX did, and often also delayed by 1.5 hours on average, if a rain event lasted 1.5 hours or longer. As such, we matched the rain data from the CO2FLX with CN measurements made right prior to rain onset, while the CN data are 1.5 hours earlier for the SMOS.

## Observational Evidences

First, by counting all individual rainfall events occurred over the 10-y period and associating them with CN measured on the ground prior to the onset of rain, we found strong, yet conditional, dependences of rain frequency on CN, as shown in Fig. 1. Distinct relationships exist for clouds with different LWP. For high LWP, the rainfall frequency increases with increasing CN but decreases for low LWP. The two variables are highly correlated for high LWP with  $R^2$  being 0.71 to 0.91 for the two rain datasets. Both regression relations are statistically significant to within a 95% confidence level. For moderate LWP, their relation appears neutral.

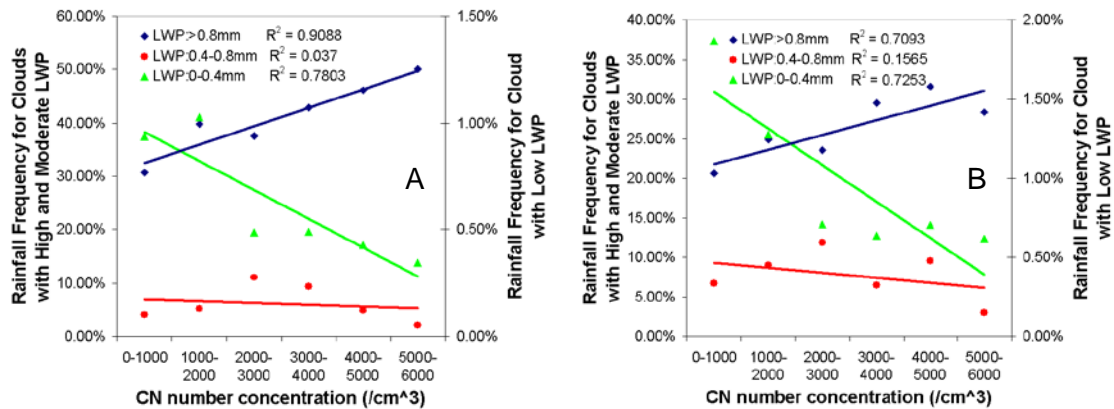


Fig. 1. Rainfall frequency as a function of CN for different LWP bins at SGP site during (A) 2003-2008 and (B) 1999-2008. Totally about 20,000 and 32,000 data points are used in the calculation of A and B, respectively. For clouds with LWP smaller than 0.4mm, the right Y axis is used to show the changes more clearly.

The relationships are not affected significantly by the selection of the CN bins, but most drastically affected by cloud base height (CBH), noting that the CN measurements were made on the ground. Fig. 2 presents relationships for two distinct ranges of cloud CBH < 1km and between 1-4 km. Rain frequency increases sharply with the CN for low CBH,

but decreases for high CBH. As elaborated below, the decrease and increase are caused by two competing effects whose strengths are dictated strongly by cloud base height. The correlation coefficient  $R^2$  is as large as 0.982 for the CO2FLX rain data, and 0.715 for the SMOS. Apparently, the low data resolution degrades the correlation.

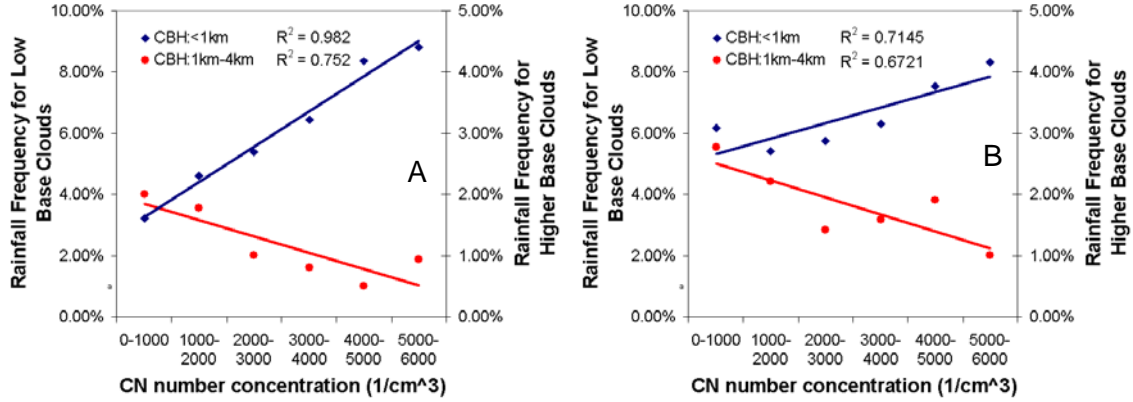


Fig. 2. Rainfall frequency as a function of CN for clouds of different cloud base heights during (A) 2003-2008 and (B) 1999-2008. The right Y-axis is for high cloud base.

Rainfall is affected by many meteorological factors such as water vapor, atmospheric stability and circulation, pressure, etc, but none was found to bear any significant connection with CN over the 10-y period (Fig. S1). This precludes the possibility that CN is a proxy of other meteorological variable(s) that could actually alter the rain frequency. A negative correlation was found between wind speed and CN due presumably to the accumulation of pollutants under light wind condition.

Given the inherent relation between rain and cloud and between LWP and cloud thickness (H), the dependence of H on CN is studied for 3 different ranges of CBH (Fig. 3). For  $CBH < 1m$ , H increases steadily and most significantly, varying by a factor of 2 or more from the cleanest to the dirtiest ensemble conditions. The trend holds for CBH 1-2km but at a lower rate. The dependence disappeared virtually for  $CBH > 2km$ . Lower CBH also implies higher temperature, higher moisture, and greater distance to the freezing level, where aerosol-induced invigoration is expected to be greater due to the release of more latent heat (5, 6), although model simulations suggest that some invigoration can also occur even without freezing (8).

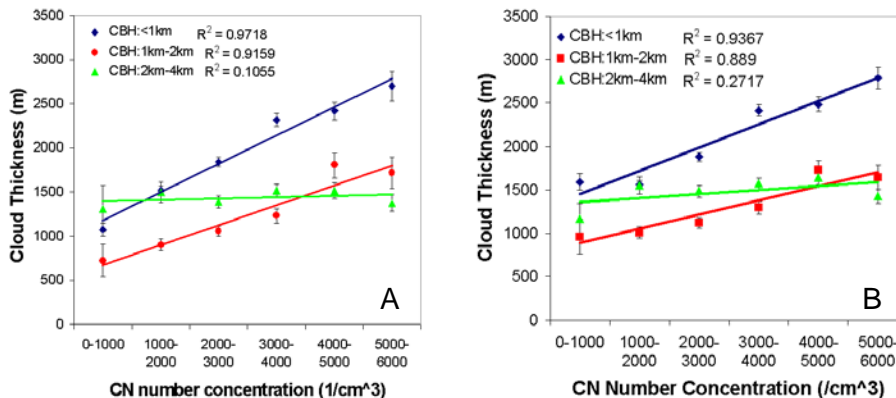


Fig. 3. Cloud thickness as a function of CN for clouds with different base heights during (A) 2003-2008 and (B) 1999-2008. The bars denote the standard differences.

The freezing-induced invigoration is confirmed by the analyses of the frequency of cloud occurrence and LWP with respect to six CN bins for six different cloud top height (CTH) ranges. As CN increases, high clouds occurred more frequently but low clouds occurred less frequently (Fig. 4a). The transition took place around the freezing level of 3.3km as determined from the 10-y ARM data. Fig. 4b reveals that cloud LWP increases drastically with the CN after it exceeds 0.8mm, the threshold used in Fig. 1, which thus explains the dependence on LWP for the relationship between rainfall frequency and CN shown in Figs. 1 and 2. Note these analyses are for  $CBH < 1\text{km}$  for which the boundary-layer aerosols have a chance to interact with clouds.

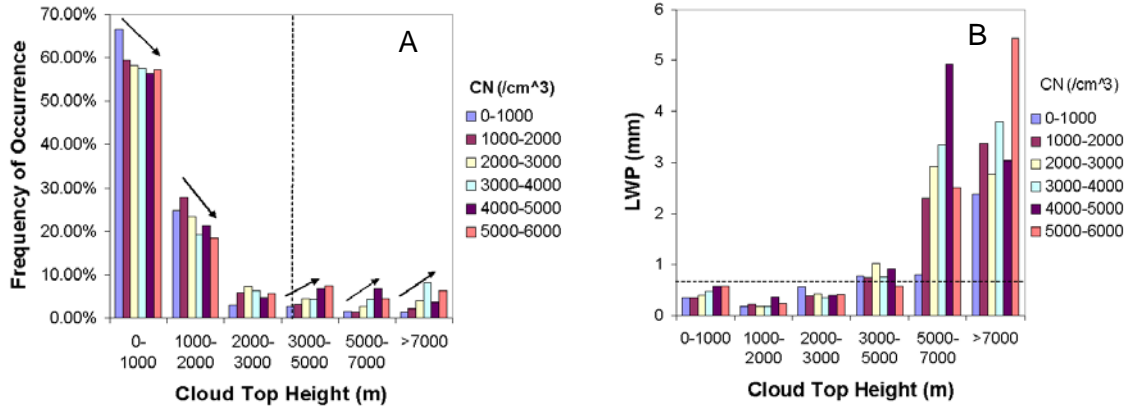
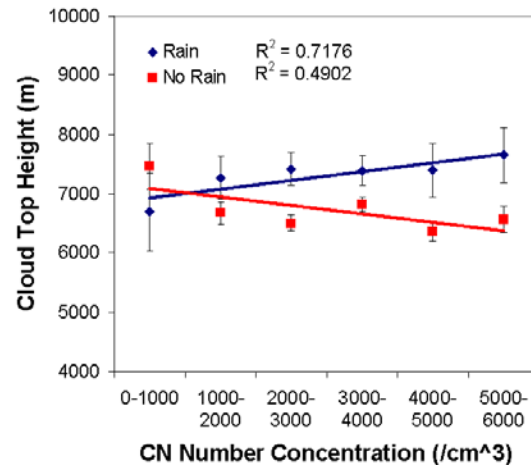


Fig. 4. (A) Frequency of occurrence of cloud top heights and (B) LWP for clouds with different top heights under different CN conditions.

The importance of ice processes in the invigoration effect as hypothesized in (6) is supported by observations as shown in Fig. 5. This figure was generated under the constraints:  $CBH < 1\text{km}$  and  $CTH > 3.3\text{ km}$  to assure interaction with aerosols and ice processes are involved. Rainy and non-rainy conditions were separated. Cloud top heights in rainy conditions were measured in the first half hour of rain. Note that the CTH increases only in rainy conditions but decreases in non-rainy conditions. This is because the

unloaded water (no rain occurrences) needs extra buoyancy to be held inside clouds. The latent heat release can just balance the weight of crystals when they reach above the freezing level. After that, only if ice particles precipitate, the cloud will be invigorated due to reduced weights and melting cooling below the freezing level (6).

Fig. 5. Changes of cloud thickness under rainy and non-rainy conditions for clouds with  $CTH > 3.3\text{km}$  and  $CBH < 1\text{km}$ .



All the aforementioned effects are expected to be strongest in the summer season when convective clouds are usually more abundant than other seasons. This is once again verified (Fig. S2-S4).

### Theoretical Interpretation

To understand the observed dependence of cloud thickness and rain frequency on aerosol concentration, we use a simple conceptual model and a full-fledged cloud resolving model to try to explain and simulate the observational findings. As stated earlier, the Twomey effect and the invigoration effect are, among others, key to the onset of precipitation. The height for the onset of precipitation ( $H^*$ ) determines partially how deep a cloud can develop.  $H^*$  is a critical parameter that increases with CN (6, 16). If cloud top height ( $H$ ) is less than  $H^*$ , rain is unlikely. But, if  $H > H^*$  and cloud is above the freezing level ( $H_0$ ), there would be an invigoration effect by freezing supercooled cloud water. If  $H^* \gg H_0$  the suppression of ice hydrometeors to greater heights would decrease the invigoration or may even lead to a suppression.

According to Figs. 1 and 4b, clouds with  $LWP < 0.4$  mm are either non convective or shallow with  $H < H^*$ , leading to the dominance of the suppression effect of increased CN. Clouds with  $LWP > 0.8$  mm are convective and deep, where invigoration occurs for  $H > H^*$  and  $H > H_0$ , where  $H_0 = 3300$  m. The cases of the mid-range LWP represent transition from negative to positive aerosol influences and thus exhibit also no dependence on CN.

$H^*$  is reached when cloud top effective radius ( $R_e$ ) exceeds a critical size  $R_e^*$  of 12 – 14  $\mu\text{m}$  (5, 17).

$$R_e = \alpha(L / N)^{1/3} \quad (1)$$

where  $L$  and  $N$  are cloud liquid water content and the cloud drop concentration respectively.

The probability of rainfall is proportional to  $R_e$  (18). When  $R_e = 15$ , a cloud has an equal chance of raining and non-raining. When  $R_e = 20$ , cloud is almost certain to rain (Fig. 8 of 18). As such, we may use  $R_e$  as a proxy of rain probability.

Note that  $L$  generally increases with height for  $H < H^*$ .  $N$  depends mainly on the CCN concentrations. Below the convective cloud base, CCN can be parameterized as a power function of aerosol number concentration ( $N_a$ ) (19).  $L$  can be parameterized as a function of  $H$  by a power law function (20, 21), we have

$$R_e = \sigma N_a^{-\alpha} H^\beta, \quad (2)$$

The two exponents,  $\alpha$  and  $\beta$ , denote the influence of aerosol number concentration and cloud thickness on cloud drop size.  $\beta$  is equal to 1/3 for an adiabatic cloud.  $H$  can be further linked to  $N_a$  based on (20):

$$H = A N_a^B \quad (3)$$

Where  $B$  denotes the strength of the invigoration effect. It follows from Fig. 3 that  $B$  is a

function of cloud base, large for clouds of low bases but near zero for clouds of high bases. Combining Eqs. (2) and (3) yields

$$R_e = CN_a^{B\beta-\alpha} \quad (4)$$

Per the magnitudes of the three coefficients, we may classify the joint effect of aerosols in three regimes  $B\beta > \alpha$ ,  $B\beta = \alpha$ , and  $B\beta < \alpha$  corresponding to strong, moderate, and weak invigoration effects relative to the strength of the rain suppression effect. These relations can be seen in Fig. S5.

In addition to the above simple model that helps explain explicitly the observation results, we attempt to simulate the observed features using a 3-D cloud-resolving model with a spectral-bin microphysics for a real thunderstorm case on Feb. 06 2006 observed from the Tropical Pacific Warm Pool International Cloud Experiment (TWP-ICE) (22). Clouds are found to be invigorated much more by lower-level aerosols than elevated-aerosols for a deep convective cloud. Simulations were conducted with a baseline CCN concentration of  $220 \text{ cm}^{-3}$  with a cloud base of about 750 m. Test runs were performed by increasing CCN to  $1320 \text{ cm}^{-3}$  in the planetary boundary layer (PBL) only and in the mid-troposphere. The modeling results are shown in Fig. 6. The former leads to a significant increase in cloud thickness, whereas little change is incurred for the latter. This is traced to changes to the updraft velocity that is increased by CCN in the PBL, but is not changed much by the mid-tropospheric CCN, indicating the invigoration of the low-base clouds by aerosols near the ground. This also support the analytical analysis in our simple model that deep convective clouds ( $LWP > 0.8 \text{ mm}$ ) are more likely to be invigorated by CCN. However, when CCN is too high, rain becomes very difficult where the rain initial height is far above the freezing level, less invigoration effect is predicted, as demonstrated in another case for extremely high CCN (i.e.,  $6600 \text{ cm}^{-3}$ ), the decrease in cloud thickness is about 1.1 km relative to the case with PBL CCN of  $1320 \text{ cm}^{-3}$ , showing the invigoration effect is significantly suppressed.

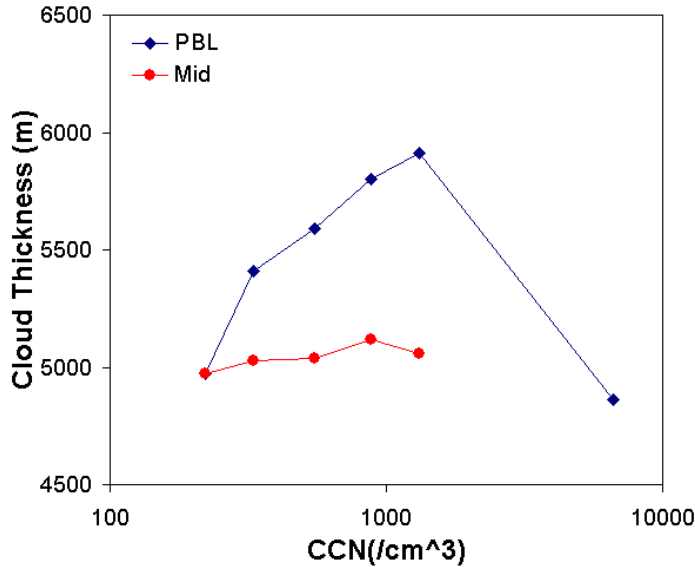


Fig. 6. Cloud thickness versus CCN concentrations in the PBL and the mid-troposphere.



## Discussion and Conclusions

Aerosol can influence cloud and precipitation in numerous ways. Most notable are the diminished coalescence (originated from the Twomey effect) and cloud invigoration effects that tend to suppress or enhance precipitation, respectively. Yet, the dominant influence of atmospheric dynamics on clouds often conceals the aerosol signals, rendering an ***unknown net effect, especially on a long time scale***. The vast majority of previous observation-based studies were concerned with individual cases.

By analyzing up to 10 years worth of measurements of aerosols, clouds and precipitation, we are able to ***reveal the net long-term effect of aerosols on clouds and precipitation and identify two dominant, co-existing yet competing effects and their determinant factors***. Their relative strengths determine how rainfall frequency responds to changes in aerosol loading. Cloud-base height plays a key role in determining if and how much aerosol interact with clouds, while cloud top height dictates the relative strength of the two effects. The largest effect occurs for deep convective clouds with low bases and high tops. Clouds with low bases usually have a high water vapor amount which fuels the invigoration effect to a degree where it outweighs the aerosol rain suppression effect. This leads to a larger cloud effective radius and to enhance rainfall. Conversely, the invigoration effect is weak in clouds with higher bases so rainfall is suppressed through the dominance of reduction in cloud effective radius by aerosols.

The observational evidence of aerosol effects on convective clouds and precipitation is a testimony to that human activities can redistribute clouds, alter precipitation and latent heating profiles to a much greater extent than have been appreciated. Incorporation of these effects in climate models may reveal that ***aerosols have a much bigger anthropogenic forcing on the climate system due to their impact on cloud thickness that has not been accounted for in the vast majority of GCMs***.

They may also have significant implications for the global hydrological cycle. One implication is that pollution can have very different influences on precipitation under different meteorological and environmental conditions over different parts of the world. Clouds over arid regions are usually shallow so in the presence of pollutants, precipitation is more likely suppressed due to the rain suppression effect, exacerbating aridity. Conversely, aerosols in moist areas are likely to deepen convective clouds to worsen flooding due to the invigoration effect. ***The strong signal of human impact on nature emerged from the long-term observations thus has tremendous social and economic consequences.***

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## Supporting online Material

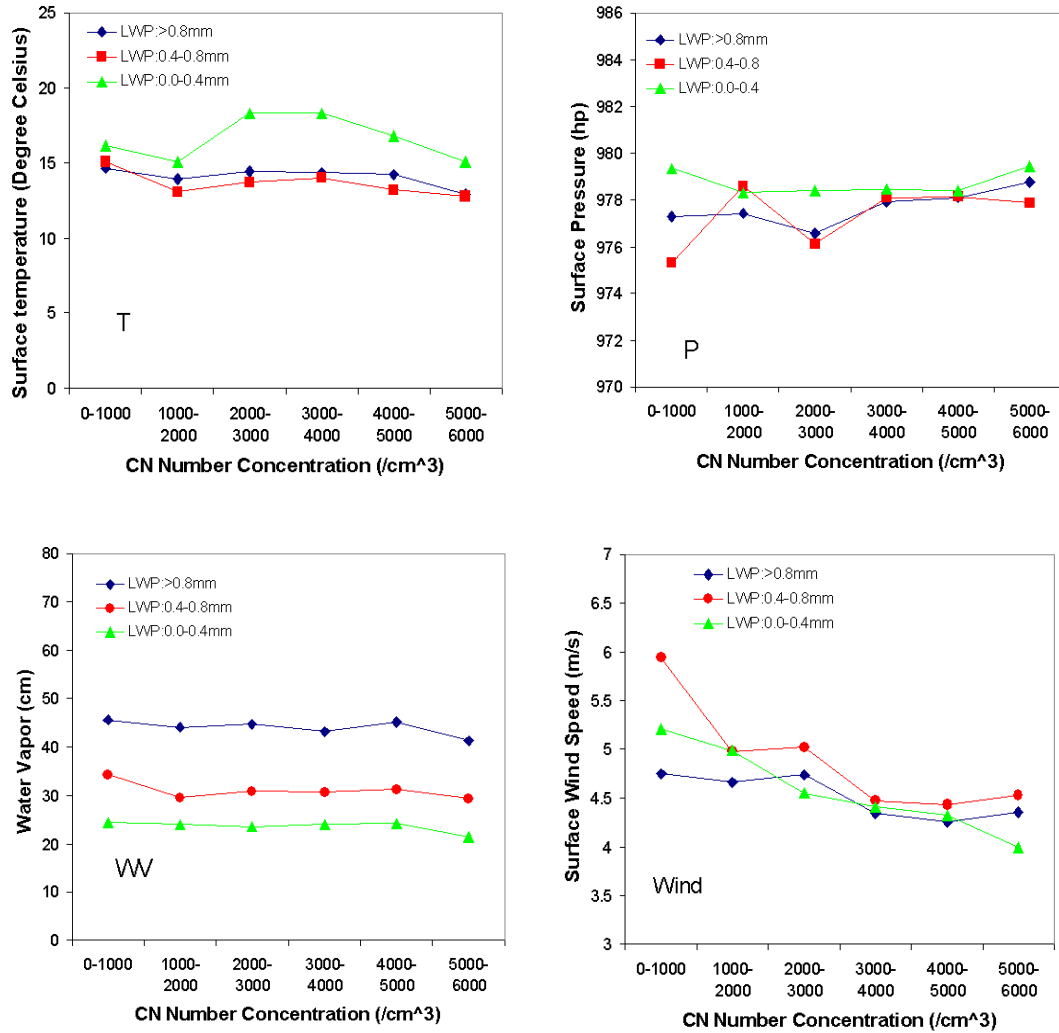


Figure 1 – Meteorological variables including surface temperature, pressure, and wind speed and column water vapor as functions of CN number concentration.

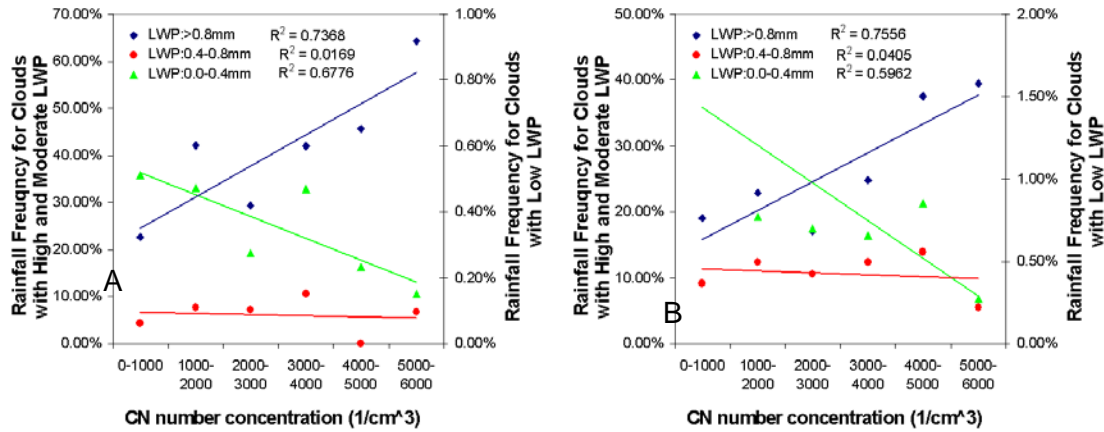


Figure 2 – Rainfall frequency as a function of CN number concentration for different LWP bins at SGP site in summer during (A) 2003-2008 and (B) 1999-2008. For clouds with LWP less than 0.4mm, the right Y axis is used to clearly show the changes.

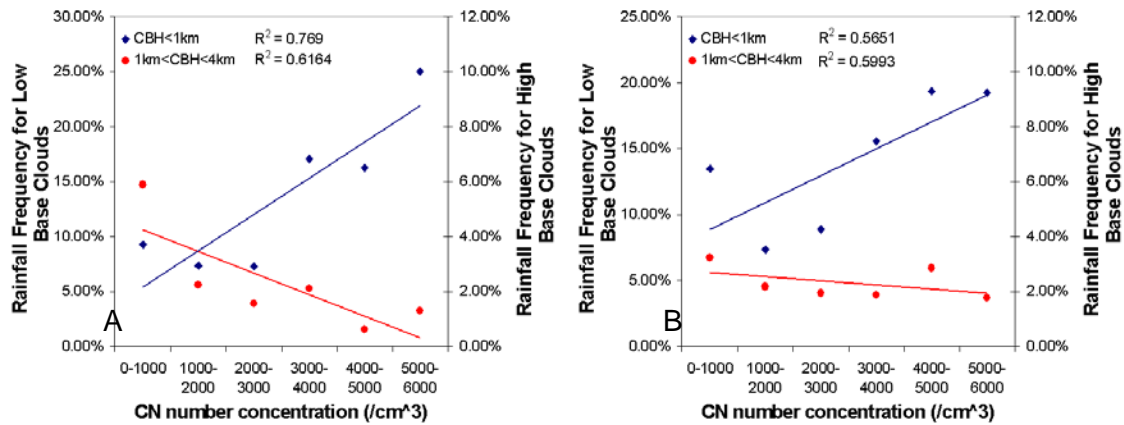


Figure 3 – Rainfall frequency as a function of CN number concentration for clouds with different cloud base heights in summer during (A) 2003-2008 and (B) 1999-2008. For clouds with higher base the right Y axis is used to clearly show the changes.

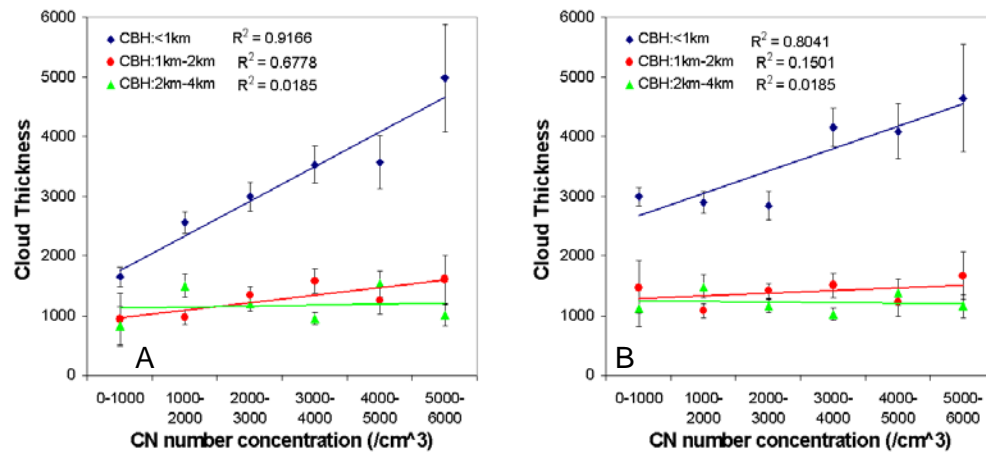


Figure 4 – Cloud thickness as a function of CN number concentration for clouds with different base heights in summer during (A) 2003-2008 and (B) 1999-2008. The error bar corresponds to the standard error of the averaged cloud thickness.

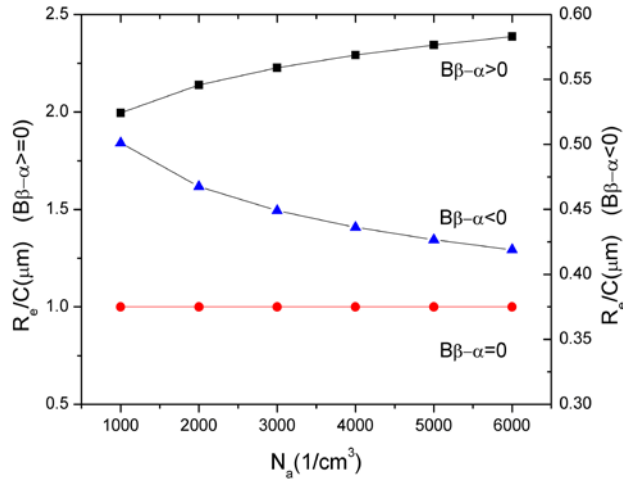


Figure 5 - A model demonstrating the competition of aerosol indirect and invigoration effects as expressed by Eq. (4). Although there is no agreement on the exact value of  $\alpha$ , the value 0.1 which is derived from the global AVHRR data (1) are used here.  $\beta$  reflects how the cloud effective radius changes with cloud thickness. The value of 1/3 for an adiabatic cloud is used. B represents the strength of the invigoration effect. From Fig.2, we see B is large for low base clouds but near zero for high base clouds. Based on above values, three different regimes  $B\beta > \alpha$ ,  $B\beta = \alpha$ , and  $B\beta < \alpha$  results in three different ranges of B:  $B > 0.3$ ,  $B = 0.3$ , and  $B < 0.3$ , respectively. The values of B 0.6, 0.3, and 0 are adopted to represent these three regimes: strong, moderate, and weak invigoration effects.

### References and Notes

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